

# Positive Magneto-Resistance in Quasi-1D Conductors

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*February 1, 2008*

We present here a simple qualitative model that interpolates between the high and low temperature properties of quasi-1D conductors. At high temperatures we argue that transport is governed by inelastic scattering whereas at low temperatures the conductance decays exponentially with the electron dephasing length. The crossover between these regimes occurs at the temperature at which the elastic and inelastic scattering times become equal. This model is shown to be in quantitative agreement with the organic conductor  $TTT_2I_{3-\delta}$ . Within this model, we also show that on the insulating side, the positive magnetoresistance of the form  $(H/T)^2$  observed in  $TTT_2I_{3-\delta}$  and other quasi-1D conductors can be explained by the role spin-flip scattering plays in the electron dephasing rate.

PACS numbers:72.10.Fk, 72.15.Nj, 75.20.Hr

At  $T = 0$ , it is well-known that disorder precludes the existence of a metallic state in strictly one and two-dimensional systems.<sup>1</sup> However, in the case of quasi-1D materials composed of coupled 1-d chains, for example, conducting polymers and organic charge-transfer salts, the situation is less clear. Abrikosov and Ryzhkin<sup>2</sup> argued that any coupling between a collection of 1-d chains will result in a non-zero conductivity at  $T = 0$ . However, based on a self-consistent diagrammatic approach, Prigodin and Firsov<sup>3,4</sup> showed that at  $T = 0$  a quasi-1D system becomes insulating for sufficiently weak interchain coupling. Specifically, they showed that only when the interchain hopping matrix element  $t$  exceeds  $0.3/\tau_{el}$ , does metallic transport obtain. Here  $\tau_{el}$  is the elastic scattering time from static impurities. There are two major reasons why this conclusion is qualitatively reasonable. First, if  $t \ll 1/\tau_{el}$ , then an electron will scatter several times before it hops onto another chain, thereby rendering interchain hopping ineffective against the localizing effect of disorder on a single chain. The second reason can be understood by considering the dispersion relationship

$$\epsilon(p) = v_F(|p_z| - p_F) + t[\cos(ap_x/\hbar) + \cos(ap_y/\hbar)] \quad (1)$$

for an anisotropic conductor, with  $v_F$  and  $p_F$  the Fermi velocity and momentum, respectively, and  $a$  the interchain spacing. We assume here that the chains form a square lattice in the  $x - y$  plane (although real lattices are almost never perfectly square or rectangular, this will prove inconsequential to our conclusions). In the limit of strong elastic scattering,  $1/\tau_{el} \gg t$ , broadening of the Fermi surface exceeds the hopping part of the dispersion relationship, thereby removing the 3-dimensional nature of the transport. In this paper we are concerned primarily with materials of this type.

In the opposite limit, when coupling between chains is relatively strong,  $t > 1/\tau_{el}$ , the material behaves as an

anisotropic 3D metal at low temperatures. In this case, the effect of quasi-one-dimensionality can be completely absorbed into a diffusion constant that is anisotropic along the crystal axes.<sup>4,5</sup> Such a material will remain conducting at the lowest temperatures, unless a structural transition such as a Peierls' distortion occurs.

In this paper, we are concerned only with materials that do not undergo Peierls' transitions. One of the outstanding problems associated with the transport properties at low temperatures is the behavior of the magneto-resistance on the insulating side. For example, in the charge transfer salt,  $TTT_2I_{3-\delta}$ ,<sup>6</sup> and the conducting polymer p-phenylenevinylene (ppv),<sup>7</sup> the magneto-resistance on the insulating side is positive and scales as  $(H/T)^2$ , with  $H$  the applied magnetic field and  $T$  the temperature. While this functional form for the magneto-resistance is generally associated with a competition between Zeeman and thermal energies, no formal account of this phenomenon has been advanced within the context of strong or weak localization. It is this problem we address in this paper. Additionally, we focus on the origin of the conductivity maximum exhibited by a wide class of quasi-1D materials, most notably  $TTT_2I_{3-\delta}$ . In  $TTT_2I_{3-\delta}$ , the conductivity displays a maximum around  $T_M = 100$  K. Above 100 K, the conductivity decreases as the temperature increases, indicating the possible onset of a metallic state. We first show that the conductivity maximum represents a crossover from strong to weak localization. At the crossover the elastic and inelastic scattering times become equal. Below  $T_M$ , we show that within a strong localization account, magnetic impurities can give rise to a positive magneto-resistance of the desired form. This effect arises from the field-dependence of the spin-flip scattering contribution to the electron dephasing rate. This explanation is in quantitative agreement with the observed experimental trends for  $TTT_2I_{3-\delta}$  as well as for (ppv).<sup>7</sup> The

agreement with (ppv) suggests that transport on the insulating side is dictated by strong localization rather than by variable-range hopping as suggested previously.<sup>7</sup>

We consider first the high temperature transport properties, that is,  $T > T_M$ . In most quasi-1D materials above  $T_M$ , the resistivity is of the form,  $\rho(T) = \rho_o + aT^\gamma$ , with  $\gamma$  anywhere between 1 and 2. The reason for the intermediate magnitude of  $\gamma$  compared to unity at high temperatures and 5 at low  $T$  for regular 3D metals is in the complex nature of the electron-phonon interaction in quasi-1D materials. This interaction is responsible for most of the temperature-dependent resistivity.<sup>6</sup> The contribution to the temperature-dependent resistivity from electron-electron interactions, on the other hand, can oftentimes be neglected as a result of the low carrier density in most of the low-dimensional conductors. Therefore, under the assumption that elastic scattering from impurities and electron-phonon scattering are independent, the high-temperature conductivity is given by the standard Drude formula

$$\sigma(T) = \frac{Nn_1e^2\tau}{m} \quad (2)$$

$$\frac{1}{\tau} = \frac{1}{\tau_{el}} + \frac{1}{\tau_{in}} \approx \frac{1}{\tau_{el}} + \frac{1}{\tau_{ph}}$$

where  $N$  is the number of chains per unit area,  $n_1 = 2p_F/\pi\hbar$  is one-dimensional density of electrons,  $m$  is the effective mass of the electrons, and  $\tau_{ph}$  is the electron-phonon scattering time.

While the low-temperature properties of strongly coupled isotropic materials can be understood in terms of weak localization,<sup>8</sup> the situation is quite different for “real” quasi-1D materials. In such materials, weak localization occurs only when the inelastic scattering rate exceeds the elastic scattering rate,  $1/\tau_{in} \gg 1/\tau_{el}$ . As a function of temperature then, a crossover is expected from the transport being governed by localization (low temperatures) to a regime in which inelastic scattering dominates (high temperatures). At low temperatures, transport is localized with a conductivity decaying exponentially<sup>9,10</sup>

$$\sigma = \frac{e^2}{\hbar} N L_\phi e^{-\frac{L_\phi}{\xi}} \quad (3)$$

with the Thouless length,  $L_\phi = v_F\sqrt{\tau_o\tau_\phi}$ . In Eq. (3),  $\xi$  is the localization length, which in 1D-conductors is on the order of the elastic mean free path,  $\ell = v_F\tau_{el}$ , and  $\tau_\phi$  is the dephasing time, which originates from all phase breaking processes. This includes all inelastic processes and also spin-flip scattering. When the concentration of magnetic impurities is small, the dephasing rate is dominated over a wide temperature range by electron-phonon scattering.

For a 1D system, the boundary separating the conductivity being given by the Drude result or the exponentially-localized Thouless form occurs when  $1/\tau_{in} \approx 1/\tau_{el}$ . Comparison of Eqs. 2 and 3 reveals that at the temperature  $T_M$ , such that  $1/\tau_\phi(T_M) = 1/\tau_{el}$ , the

high and low-temperature conductivities predict a maximum value of

$$\sigma_{\max} \simeq \frac{e^2}{\hbar} N v_F \tau_{el}. \quad (4)$$

Above and below  $T_M$ , the conductivities can be expressed in terms of  $\sigma_{\max}$  and the temperature-dependent ratio  $\tau_\phi(T)/\tau_{el}$ . In particular, if we assume that in the wide range of temperatures, which includes  $T_M$ ,  $1/\tau_\phi(T) \propto T^\gamma$ , then

$$\sigma(T) = \begin{cases} \sigma_{\max} \left(\frac{T_M}{T}\right)^{\frac{\gamma}{2}} e^{1-(T_M/T)^{\frac{\gamma}{2}}}, & \text{if } T < T_M \\ \frac{2\sigma_{\max}}{1+(T_M/T)^\gamma}, & \text{if } T > T_M \end{cases} \quad (5)$$

Despite the simple considerations that led to Eq. (5), this expression is of extreme utility in understanding the nature of the maximum in the conductivity. The exponent  $\gamma$  can be easily extracted from the experimental data. If the ratio of the conductivity maximum to the room temperature conductivity is  $\sigma_{\max}/\sigma_{RT} \simeq (T_M/T)^\gamma/2$ , then it is likely that the observed transition is of the type described here. In fact, this condition is satisfied for the  $(TTT_2I_{3-\delta})$  samples studied by Khanna, et. al.<sup>6</sup> In their samples the conductivity maximum occurred at  $T_M \approx 100$  K and the thermal exponent  $\gamma$  is approximately 2.<sup>11</sup> Within our picture this yields  $\sigma_{\max}/\sigma_{RT} \sim 4$ , which is reasonably close to the observed ratio of 2–3. At low temperatures, the conductivity declines rapidly which is consistent with the first of the expressions in Eq. (5). However, at very low temperatures, the deviation from the predicted behavior becomes significant. For example, at  $T = 4$  K conductivity is roughly 1 to 10 percent (depending on the degree of disorder) of  $\sigma_{RT}$ . Although this value is small, it is significantly larger than that predicted from the first of the expressions in Eq. (5). The source of this discrepancy is straightforward to pinpoint. In obtaining Eq. (5) we used that  $1/\tau_{in}(T) \propto T^\gamma$ , which is essentially an extrapolation of the high temperature electron-phonon scattering rate to low temperatures, which can be expected to break down, for instance, when the thermal phonon wavelength,  $\lambda_{ph}$ , becomes comparable to the mean free path (dirty limit). Even more importantly, up to this point, we have neglected all other dephasing mechanisms besides electron-phonon scattering. As the temperature decreases, the relative role of the electron-electron interaction increases. Also, spin-flip scattering has not been incorporated. In fact, transport measurements reveal the presence of a positive magnetoresistance of the form  $(H/T)^2$  below 20 K.<sup>6</sup> We now show that inclusion of spin-flip scattering is essential to explaining the positive magnetoresistance. Hence, this effect influences the conductivity as well.

It is well known that the spin-flip scattering suppresses localization.<sup>12</sup> Unlike electron-phonon scattering, spin-flip scattering is essentially temperature-independent in the absence of a magnetic field for temperatures higher than the Kondo temperature,  $T_K$ .<sup>13</sup> Below  $T_K$  the spin-flip scattering rate gradually decreases and disappears

completely at  $T = 0$  for spin  $S = 1/2$  impurities. When spin-flip scattering is no longer negligible compared with the other (inelastic) dephasing processes, we must explicitly include it in the dephasing rate:

$$\frac{1}{\tau_\phi} = \frac{1}{\tau_{\text{in}}} + \frac{1}{\tau_s} \quad (6)$$

where  $\tau_s$  is the spin-flip scattering time and  $\tau_{\text{in}}$  now represents all inelastic processes, such as electron-electron interactions and electron-phonon scattering.

Of these processes, the only one that couples to a magnetic field is spin-flip scattering. In a non-magnetic 1D system, the fact that flux cannot be enclosed signals a vanishing of the localization correction to the magnetoresistance. Hence, magnetic impurities offer a channel to circumvent the apparent lack of magnetic coupling in 1D systems. In the presence of a field, the spin-flip scattering rate decreases because the spins partially align with the field. As a consequence,  $L_\phi$  grows and the conductivity given by the first of the Eqs. (5) decreases. Hence, the magnetoresistance is positive. To determine the functional form of the magnetoresistance, we calculate the spin-flip scattering time in the presence of the field. To second order in the exchange interaction, the spin-flip scattering rate is given by<sup>14</sup>

$$\frac{1}{\tau_s} = \frac{1}{\tau_s^o} \left( \frac{1}{2} + \frac{2\beta h}{\sinh(2\beta h)} \right) \quad (7)$$

where  $\tau_s^o$  is the spin-flip scattering time in the absence of magnetic field,  $\beta = 1/(k_B T)$  and  $h = g\mu_B H/2$ . Here  $\mu_B$  is the Bohr magneton and  $H$  is the applied magnetic field. In the experimental range of interest  $\beta h < 1$ . Consequently, we expand  $\sinh x$  to obtain,

$$\frac{1}{\tau_s} = \frac{1}{\tau_s^o} \left( \frac{3}{2} - \frac{2}{3}(\beta h)^2 \right) \quad (8)$$

Hence, the spin-flip scattering rate decreases as  $(H/T)^2$ . If we simply insert this expression into the dephasing length, we find immediately that in the leading power of  $H/T$ , the magnetoconductance is

$$\frac{\sigma(H) - \sigma(0)}{\sigma(0)} = -\sqrt{\frac{\tau_\phi^3}{\tau_{\text{el}}\tau_s^o}} \left( \frac{\mu_B H}{3k_B T} \right)^2 \quad (9)$$

In general,  $\tau_\phi$  is a function of temperature since all inelastic processes are temperature-dependent. But for low enough temperature (but still higher than  $T_K$ !), spin-flip scattering can become the dominant dephasing mechanism, and then from  $\tau_\phi \approx \tau_s$  and equation (9) it immediately follows that the magnetoconductance

$$\frac{\sigma(H) - \sigma(0)}{\sigma(0)} = -\sqrt{\frac{\tau_s^o}{\tau_{\text{el}}}} \left( \frac{\mu_B H}{3k_B T} \right)^2 \quad (10)$$

is of the  $(H/T)^2$  form often seen experimentally. In particular, in the experiment of Khanna and coworkers<sup>6</sup> such

a positive magnetoresistance was observed below 20 K. No anisotropy was observed, which strongly supports the spin nature of the magnetoresistance. If we assume that the dominant dephasing mechanism is given by electron-phonon scattering, we find that  $\tau_\phi \approx \tau_{\text{el}}(T/T_M)^\gamma$ . From this we estimate that the spin-flip scattering time  $\tau_s \approx \tau_\phi(20K) \approx 25\tau_{\text{el}}$ . Applying Eq. (10) then gives  $\Delta\sigma(H)/\sigma(0) \simeq 0.2(H/T)^2$ , where  $H$  is in Tesla and  $T$  in Kelvin. The corresponding prefactor extracted from the experimental data<sup>6</sup> is between 0.1 and 0.2 which is in excellent agreement with our qualitative prediction. At even lower temperatures (below 4 K) the field and temperature dependence of the magnetoresistance become weaker. At the same time, the zero-field conductivity continues to decrease. These observations are consistent with the quenching of the impurity spin by the conduction electrons below the Kondo temperature,  $T_K$ .

We have shown quite generally that quasi-1D systems in which localization effects are dominant acquire a positive magneto-resistance of the form  $(H/T)^2$  through spin-flip scattering from magnetic impurities. This correction was shown to be in quantitative agreement with the experiments on the charge transfer salt  $TTTT_2I_{3-\delta}$ . A similar positive magneto-resistance was observed in the conducting polymer ppv.<sup>7</sup> Heeger and co-workers<sup>7</sup> found that neither the Mott variable-range hopping model<sup>15</sup> nor the Efros-Shklovskii Coulomb gap model<sup>16</sup> could account for the  $(H/T)^2$  trend in the magneto-resistance. The model presented here offers a natural explanation of this experimental trend. Experimental susceptibility measurements on ppv could be used to test the presence of magnetic impurities and hence confirm the scenario presented for the origin of the positive magneto-resistance.

## ACKNOWLEDGMENTS

This work was supported by the NSF grant No. DMR94-96134 and the donors of the Petroleum Research Fund administered by the American Chemical Society.

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